

Production of ^{148}Gd by 600- and 800-MeV Protons on Tungsten, Tantalum, and Gold

LA-UR-03-056

R.K. Corzine (LANSCCE Division and Georgia Institute of Technology), M.J. Devlin, E.J. Pitcher (LANSCCE Division), N.E. Hertel (Georgia Institute of Technology), S.G. Mashnik (X and T Divisions)

A

t LANSCE, protons are accelerated to 800 MeV and directed to two tungsten targets — Target 4 at the WNR and the Mark 1 Target at the Lujan Center. A cascade of radioisotopes are produced by spallation reactions in these targets. The Department of Energy (DOE) requires hazard classification analyses performed on these targets and places limits on certain radionuclide inventories that are produced. These analyses help the facilities to be kept below the threshold that characterizes nuclear facilities.

^{148}Gd is one such radioisotope created from spallation reactions in a tungsten target. Allowed isotopic inventories are particularly low for this radioisotope because it is an α -particle emitter with a 75-year half-life. The activity level of ^{148}Gd is low; however, based on present yield estimates, it amounts to almost two-thirds of the total dose burden for the two tungsten targets at LANSCE. From a hazard-classification standpoint, this severely limits the time these tungsten targets can be used. The production cross section for ^{148}Gd is not well known, hence, the motivation for measuring it.

A Need for Better Knowledge of the ^{148}Gd Production Rates in Tungsten Targets in the 600- to 800-MeV Energy Range

The 800-MeV protons lose about 200 MeV when passing through the tungsten targets at WNR and the Lujan Center. They therefore exit the targets with an energy of 600 MeV. Because DOE limits the inventory of ^{148}Gd , a better estimate of the ^{148}Gd production rate in tungsten targets is needed for proton energies between 600 MeV and 800 MeV. From a basic

nuclear-physics standpoint, the ideal strategy would be to measure the production cross sections for each tungsten isotope in this energy range. However, obtaining isotopically pure tungsten foils is cost prohibitive. An alternative is to perform measurements with a mono-isotopic element close to that of tungsten ($Z = 74$) in atomic number. Tantalum ($Z = 73$), which is 99.988% ^{181}Ta , provides a good alternative for testing the physics models used to estimate spallation products at these energies. Furthermore, tantalum is used as

target cladding at two spallation-neutron-source (SNS) facilities — KENS (Japan) and ISIS (Rutherford-Appleton Laboratory Neutron Facility, United Kingdom) — which operate at 500 MeV and 800 MeV, respectively. By measuring ^{148}Gd production from tantalum, we can use nuclear-physics models with measured production cross sections from elemental tungsten to gain a better understanding of production rates for individual tungsten isotopes. These models will then help in evaluating dose burdens at other SNS facilities.

Several other measurements of spallation-product yields exist for intermediate-energy proton interactions with tungsten, tantalum, and gold. Gold, a mono-isotopic element ($Z = 79$), is of interest to the new Spallation Neutron Source located at Oak Ridge National Laboratory because of its atomic-number proximity to that of mercury ($Z = 80$), the candidate target material. Previous measurements^{1,2,3} used γ -ray spectroscopy, radiochemical analysis, or a fragment separator to determine independent and cumulative (which includes decay from radioactive parents) spallation-product yields.

The ^{148}Gd inventory is difficult to deduce in a thick target because it decays only by α -particle emission (with a short range) with no associated γ -ray emission. To date, only one measurement of the number of ^{148}Gd atoms produced from tungsten has been made. A radiochemistry analysis, which was done as part of the decay-heat experiment for the Accelerator Production of Tritium (APT) project, measured the number of ^{148}Gd atoms in the center of three tungsten foils irradiated with 800-MeV protons.¹ Assuming that this radioisotope is only produced within the confines of the beam spot, a ^{148}Gd production cross section of 16.40 ± 0.41 mb can be inferred from this measurement. A current theoretical estimate obtained with the CEM2k+GEM2 code⁴ for the cumulative production of ^{148}Gd from ^{184}W is 24.24 ± 0.19 mb. For purposes of hazard classification, conservative efforts require that this theoretical estimate be multiplied by a factor of 2, which further limits the time that the target can be irradiated.

To assess the production of ^{148}Gd in a thick target, we performed a series of thin-foil irradiations. The foils had to be thin enough for the α -particles from ^{148}Gd decay to exit the foils and be detected. The acceptable foil thickness was 3 μm for tungsten, tantalum, and gold. Aluminum foils

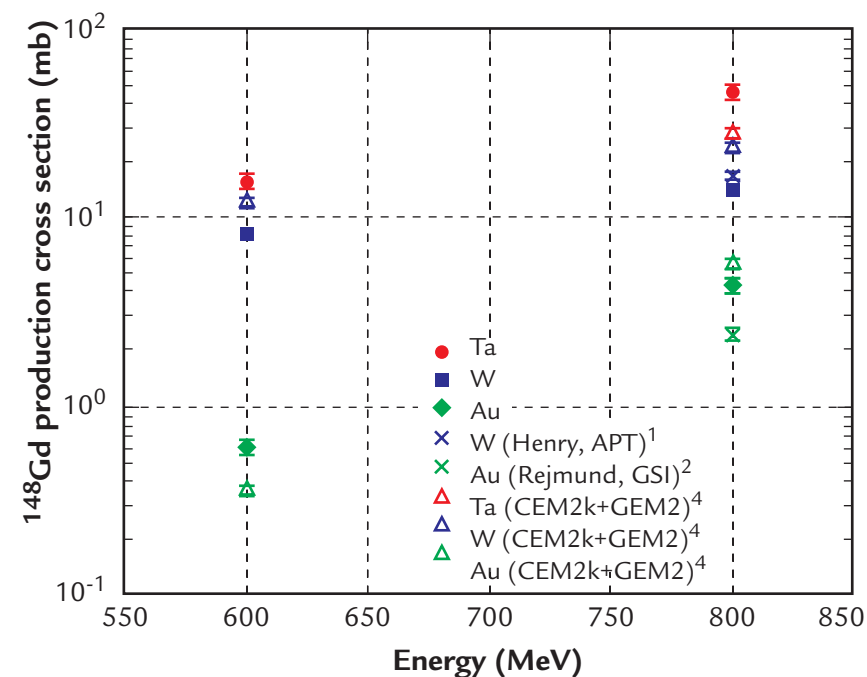


Fig. 1. Preliminary ^{148}Gd -cumulative-production-cross-section measurements for tungsten, tantalum, and gold interactions with 600- and 800-MeV protons at WNR compared to previous measurements and predictions.

(10 μm) were used to determine an absolute proton flux on the foils using the well-known $^{27}\text{Al}(p,x)^{22}\text{Na}$ cross section. Stacks of three foils were used to investigate any possible loss of ^{148}Gd and ^{22}Na recoils in the material of interest. ^{148}Gd cross sections were determined by directly counting decay α -particles with the appropriate energy in a dedicated detector system.

During the 2002 run cycle at LANSCE, we irradiated thin samples of tungsten, tantalum, gold, and aluminum with 600- and 800-MeV protons at WNR to obtain cross-section measurements and compare them to previous measurements and theoretical estimates from the CEM2k+GEM2 code (Fig. 1). Preliminary analyses indicate that our measurements for tungsten are within 40% of CEM2k+GEM2 predictions and within 14% of the APT decay-heat measurement. Another set of irradiations at 800 MeV is planned for January 2003

in the WNR Blue Room. Measurements for other isotope-production cross sections are also being investigated via γ -ray spectroscopy and compared to previous measurements and predictions.

Acknowledgements

The authors would like to acknowledge the operators in the Central Control Room (LANSCE-6) and the support of Gregg Chaparro (LANSCE-3) in the WNR Blue Room. This study is supported by the U.S. Department of Energy.

References

1. E.A. Henry and K.J. Moody, APT internal report, Lawrence Livermore National Laboratory (1999).
2. F. Rejmund et al., *Nuclear Physics A* **683**, 540-565 (2001).
3. R. Michel et al., *Nuclear Instruments and Methods B* **129**, 153-193 (1997) and references therein.
4. S.G. Mashnik et al., Los Alamos National Laboratory report LA-UR-02-0608 (2002).